Implications of $d_{x^2-y^2}$ symmetry and faceting for the transport properties of grain boundaries in high- T_c superconductors

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Grain boundaries in high- T_c superconductors have attracted wide interest for their potential in a variety of applications and in fundamental studies of high- T_c superconductivity. Two recent experimental results provide a basis for a better understanding of the grain boundary properties, the mechanisms of which, despite their widespread use, are not yet completely understood. First, it is now well established that the order parameter in many high- T_c cuprates has a predominant $d_{x^2-y^2}$ symmetry. Second, microscopy studies have revealed that practical grain boundaries are comprised of facets having various orientations and typical dimensions of the order of 10–100 nm. We analyze the combined effects of faceting and $d_{x^2-y^2}$ symmetry on the transport properties of high- T_c grain boundaries. It is found that these effects can partially account for the experimentally observed reduction of the critical current density J_c with increasing grain boundary angle α . The angular dependence of J_c for individual grain boundary facets may deviate considerably from the $J_c(\alpha)$ dependence of J_c and the normal state resistivity ρ_n . The $J_c\rho_n$ product measured for standard grain boundary junctions is therefore not a direct measure of the intrinsic barrier properties. The faceting and $d_{x^2-y^2}$ symmetry lead to an inhomogeneous current distribution in the grain boundary which is different for the superconducting and the normal states. [S0163-1829(96)05521-X]

Grain boundaries are extremely important for applications and for basic investigations of high- T_c superconductors. They are used as high-quality Josephson junctions in a variety of device applications. Owing to their critical currentlimiting properties, they are also the focus of research on large current applications of high- T_c superconductors. Furthermore, their unique properties provide insight into the fundamentals of high- T_c superconductivity, such as the symmetry of the superconducting order parameter.^{1–3} Despite extensive research conducted on high- T_c grain boundaries, the mechanisms determining their behavior are still under debate and a complete quantitative understanding has not yet been achieved.

Recently, two properties of the high- T_c cuprates, highly relevant for current transport across grain boundaries, have become established experimental facts. First, several experiments provided clear evidence that the symmetry of the order parameter of many high- T_c superconductors is $d_{x^2-y^2}$ (Refs. 1,3-8) or $d_{x^2-y^2}$ mixed with an s-wave component.⁹ Here x and y denote the directions of the \vec{a} and \vec{b} axes of the cuprates, or vice versa. Second, transmission electron microscopy (TEM) has disclosed that thin-film grain boundaries are generally composed of facets having typical dimensions of the order of 10-100 nm.¹⁰⁻¹⁷ The facet orientations and lengths depend on the grain boundary configuration, on the high- T_c material, the substrate, the conditions used for film deposition, and the presence of defects. Faceting, which takes place in all three dimensions, is a consequence of the growth modes of the cuprates and is observed for grain boundaries fabricated by the usual techniques employing bicrystals, biepitaxial growth, or step edges. Here, we shall attempt to clarify the implications of $d_{x^2-y^2}$ symmetry and faceting for the transport properties of the grain boundaries. The mechanisms to be described are not only relevant for high- T_c grain boundary junctions, but, possibly in a modified form, for other junctions as well, such as for superconducting-normal-superconducting (SNS) step edge¹⁸ and ramp-type devices¹⁹ or for junctions written by direct electron beam^{20,21} or ion beam irradiation.²²

The electronic behavior of high- T_c grain boundaries depends on the misorientation of the two grains.²³ A strong decrease of the critical current density J_c with increasing misorientation angle α was revealed by Dimos *et al.*^{23,24} and Ivanov et al.25 To illustrate this decrease, a compilation of $J_c(\alpha)$ data of YBa₂Cu₃O_{7-x} boundaries, extracted from Refs. 24–47 is presented in Fig. 1 (T=4.2 K). Although there is substantial scatter in the data, which partly reflects differences in film quality, it is obvious that J_c drops strongly (over more than three decades) as α is increased from 0° to 45° . To a first approximation, this decrease is exponential: $J_c \approx 2 \times 10^7 e^{(-0.18\alpha)}$ A cm⁻² (α in degrees). Symmetric and asymmetric, [001] tilt, [100] tilt, and [001] twist boundaries apparently show the same behavior. This suggests that the exponential drop of J_c is independent of the grain boundary type.²⁴

Grain boundaries with $\alpha \ge 8^{\circ}$ are known to behave like strongly coupled Josephson junctions.^{24,48} The product of their critical current density J_c and their normal state resistivity ρ_n drops with increasing misorientation α . In Fig. 2(a), $J_c\rho_n$ is plotted as a function of α for those samples of Fig. 1 for which the $J_c\rho_n$ value was reported. As a function of critical current density, the $J_c\rho_n$ product seems to vary approximately as $(J_c)^{0.6}$, as noted by Russek *et al.*²⁶ and Gross *et al.*²⁷ and shown in Fig. 2(b).

Several mechanisms have been suggested to limit the current flow across high- T_c grain boundaries.⁴⁹ Clearly, a reduc-

14 586



FIG. 1. Experimentally observed dependence of the critical current density J_c of thin-film grain boundary junctions on the misorientation angle α (T=4.2 K). The data have been compiled from Refs. 24–27. In cases where only the value at T=77 K was reported, the value expected at T=4.2 K has been extrapolated using the temperature dependence of J_c described in Ref. 48. A distinction has been made between symmetric [001] tilt boundaries ($\alpha_1 = \alpha_2 = \alpha/2$) (solid squares), asymmetric [001] tilt boundaries ($\alpha_1 \neq \alpha_2$, one of these angles usually being 0°) (open circles), symmetric [100] tilt boundaries (upright triangles), symmetric [001] twist boundaries (inverted triangles), grain boundaries in polycrystalline films (diamonds), and basal plane tilt grain boundaries (solid circles).

tion of the order parameter caused by structural disorder associated with the boundary 50,51 or by deviations from bulk stoichiometry in the vicinity of the grain boundary will reduce J_c .⁵² It was also realized very early⁴⁹ that a nons-wave-type symmetry of the superconducting order parameter would provoke an angle-dependent J_c . However, as the observed $J_c(\alpha)$ dependences for grain boundaries with different types of misorientations did not show the expected disparate behavior, it was concluded that this is not the dominant mechanism controlling J_c .⁴⁹ In the evaluation of the angular dependence of J_c the implications of a $d_{x^2-y^2}$ symmetry of the superconducting order parameter have, to our knowledge, never been considered together with the faceted microstructure of the grain boundary. Here, we attempt to assess these contributions to the observed $J_c(\alpha)$ dependence and identify additional consequences for the behavior of grain boundaries in high- T_c superconductors.

Current transport across a high- T_c grain boundary in the superconducting state depends on the order parameters of both grains close to the interface (see, e.g., Ref. 53) and, if the order parameters are anisotropic, on their orientations. The following expression for the dependence of J_c on the orientations of the order parameters has been given by Sigrist and Rice:^{54,55}



FIG. 2. Experimentally observed dependence of the $J_c\rho_n$ product on (a) the misorientation angle α (T=4.2 K) and (b) J_c , for those samples of Fig. 1 for which $J_c\rho_n$ was reported (T=4.2 K). In cases where only the value at T=77 K was reported, the value expected at T=4.2 K has been extrapolated.

$$J_{c} \propto \chi_{1}(\vec{n}_{1}) \chi_{2}(\vec{n}_{2}).$$
 (1)

The vector \vec{n}_m indicates the orientation of the interface normal with respect to the crystal lattice on side m. The $\chi(\vec{n})$ functions account for the symmetry properties of the order parameters and for their orientations with respect to the boundary. In principle, $\chi(\vec{n})$ can be extended to include all factors that influence J_c and depend on \vec{n}_m . An expression for $\chi(\vec{n})$ is yet to be experimentally established for the various high- T_c superconductors. In search of a first ansatz for a superconductor with $d_{x^2-y^2}$ symmetry, we follow Sigrist and Rice to obtain

$$\chi(\vec{n}_m) = n_{m,x}^2 - n_{m,y}^2, \qquad (2)$$

where $n_{m,x}$ and $n_{m,y}$ are the components of n_m in the crystal basis on side *m*. We emphasize that Eq. (2) takes only current flow parallel to \vec{n} into account. Accounting in addition for off-directional current components would enhance the



angular dependence of $\chi(n)$. It may be argued that the presence of the grain boundary disturbs the symmetry properties of the grains close to the boundary. It is assumed, however, that this is not a dominating effect because $d_{x^2-y^2}$ symmetry was found in several experiments, only some of which involved grain boundary junctions. One could also imagine, because orthorhombic high- T_c cuprates are usually twinned, that the sign of the order parameter varies from twin domain to twin domain. For energy reasons, however, the order parameter has only one orientation across the entire grain, as shown by tricrystal ring experiments.¹ Therefore we will not differentiate between the \vec{a} and \vec{b} axes of the cuprate superconductors.

For clarity, we will focus in the following on [001] tilt boundaries, which are the ones most commonly used. For this configuration, Eqs. (1) and (2) yield

$$J_c \propto \prod_{m=1,2} \left[(\sin \alpha_m)^2 - (\cos \alpha_m)^2 \right].$$
(3)

Here, α_m (m=1,2) is the smallest angle between the grain boundary plane and a principal axis (\vec{a} or \vec{b}) of grain m, as sketched in Fig. 3.

In Fig. 4(a), the $J_c(\alpha)$ dependence following from Eq. (3) is presented for straight, symmetric boundaries (α_1 $= \alpha_2 = \alpha/2$) and for completely asymmetric boundaries $(\alpha_1 = \alpha, \alpha_2 = 0^\circ)$. For a given angle $\alpha \leq 45^\circ$ the symmetric boundaries are those with the highest J_c . Figure 4(b) shows the same calculations on a logarithmic scale. Based on these calculations, an appreciable difference in the experimentally observed angular dependence of J_c between symmetric and asymmetric boundaries is expected. Also shown in Figs. 4(a) and 4(b) is the experimentally observed $J_c(\alpha)$ obtained from Fig. 1. It is evident that $d_{x^2-y^2}$ symmetry by itself can account for only a minor part of the experimentally observed reduction of J_c , in particular for symmetric grain boundaries. For instance, for the widely used symmetric 24° and 36.8° boundaries reductions of 17% and 36% are derived from Eq. (3), whereas the experimentally observed drops are about two and three orders of magnitude, respectively. For asymmetric boundaries the calculated reductions are 33% and 72%, respectively.

For these calculations, the grain boundaries are assumed to be straight. As mentioned above, practical grain boundary junctions consist of facets with various orientations. We will show below that the faceting strongly enhances the influence of $d_{x^2-y^2}$ symmetry on the angular dependence of J_c . For the sake of clarity we will first consider only the faceting in the plane parallel to the substrate surface and omit the influences of the boundary plane tilt with respect to the substrate normal.

FIG. 3. Schematic top view of a high- T_c grain boundary. The magnification indicates the face-

ting of the grain boundary and the angle α_f be-

tween the facet and the macroscopic boundary

As we restrict our discussion to the effects of $d_{x^2-y^2}$ symmetry on J_c , Eqs. (1)–(3) may provide an appropriate starting point to describe the individual facets. With this and with the knowledge of the facet orientations provided by TEM studies the local critical current density $J_c(\vec{r})$ can be evaluated. Figures 5(a)–5(d) show the faceted boundary lines of various grain boundaries on bicrystalline substrates extracted from TEM studies reported in the literature.^{13,15,16} Also shown are the corresponding $J_c(\vec{r})$ dependences derived from Eq. (3).

plane.

Obviously, the faceting together with $d_{x^2-y^2}$ symmetry causes the critical current density to be very inhomogeneous



FIG. 4. (a) Angular dependence of the critical current density J_c calculated according to Eq. (3) for the symmetric boundary ($\alpha_1 = \alpha_2 = \alpha/2$) and the completely asymmetric boundary ($\alpha_1 = \alpha$, $\alpha_2 = 0^\circ$). Also indicated are the experimentally observed dependence and the angular dependence of J_c attributed to the combined effects of $d_{x^2-y^2}$ symmetry and faceting. (b) Same as (a) on a logarithmic scale.

J_c/J_c (0°)

(b)

500 nm







FIG. 5. Faceted grain boundary line extracted from TEM studies reported in the literature and corresponding spatial distribution of J_c according to Eq. (3) and neglecting microstructural properties. (a) Symmetric 23° (Ref. 16), (b) symmetric 36° (Ref. 15), (c) symmetric 38° (Ref. 16), (d) symmetric 67° (Ref. 13), equals asymmetric 23° (α_1 =33.5°, α_2 =56.5°). In these figures, *r* denotes the position along the grain boundary, following the meandering path of the boundary. The length scales indicated apply for the depicted faceted grain boundary line as well as for the calculated spatial distribution of J_c . (e) Fractional area of facets with J_c =0 (open circles) and of π facets (solid squares) for the grain boundaries analyzed here.

across the boundary. It has been reported in several studies that grain boundaries on bicrystalline substrates consist largely of facets corresponding to the bicrystal misorientation and of facets that correspond to a low-index plane of one of the grains,^{12,13,15,16} such as the facets oriented parallel to the (110) plane of one of the grains ($|\alpha_1|=45^\circ$, $|\alpha_2|=|\alpha-45^\circ|$), which according to Eqs. (1)–(3) have a zero critical current, and the completely asymmetric facets

oriented parallel to the (100) plane of one of the grains $(\alpha_1 = \alpha, \alpha_2 = 0^\circ)$. As mentioned above and shown in Figs. 4(a) and 4(b), these facets have a low J_c , particularly for large α .

Furthermore, it is a striking consequence of $d_{x^2-y^2}$ symmetry that for facets with

$$|\alpha_1 - \alpha_f(\vec{r})| < 45^\circ < |\alpha_2 + \alpha_f(\vec{r})|$$

or $|\alpha_2 + \alpha_f(\vec{r})| < 45^\circ < |\alpha_1 - \alpha_f(\vec{r})|,$ (4)

the sign of J_c is opposite to the sign of J_c for facets that do not comply with this condition. In other words, in the absence of magnetic flux in the boundary, the phase differences of the superconducting order parameters over the boundary differ by π for these different facet types. In Eq. (4), $\alpha_f(\vec{r})$ is the angle between the facet and the macroscopic boundary plane illustrated in Fig. 3.

The order parameters of the grains will adjust their signs such that the total critical current of the grain boundary junction will be positive. In the following, we will refer to facets with positive critical current densities as 0 facets and those with negative J_c values as π facets. Typically, the 0 facets correspond to the facets that comply with Eq. (4); an exception is shown in Fig. 5(d) for a symmetric 67° grain boundary.

The negative J_c values of the π facets efficiently reduce the total critical current of the grain boundary. A considerable decrease of J_c with increasing α is attributed to this mechanism. Whereas for the symmetrical 23° grain boundaries only a few π facets can be identified [less than 5% of the junction area; see Fig. 5(a)], for the symmetric 36° grain boundaries the fraction of π facets can be as high as 20%; see Fig. 5(b). The fractional area of facets with $J_c=0$ and of π facets is indicated as a function of α in Fig. 5(e) for the grain boundaries analyzed here.

A spatially inhomogeneous critical current density in grain boundary junctions, on the length scales discussed here, has been proposed by various groups; see, e.g., Refs. 26,56–59. Above, we have shown that the inhomogeneity results naturally from $d_{x^2-y^2}$ symmetry and faceting, even leading to regions with negative critical current densities in the junction.

Faceting also leads to an increase of the effective grain boundary area, which is expected to enhance the critical current. This increase is estimated to be several tens of a percent and is not expected to play an important role in the angular dependence of J_c . It is to be noted that for standard \vec{c} -axis-oriented films the grain boundary meandering in the \vec{c} direction will lead to a Josephson current parallel to the \vec{c} direction, for which Eq. (3) does not apply.

With the $J_c(r)$ dependences presented in Figs. 5(a)–5(d), the contribution of the combined effects of faceting and $d_{x^2-y^2}$ symmetry to the experimentally observed angular dependence of J_c can be estimated. The resulting angle dependence of J_c ascribed to these combined effects is displayed in Figs. 4(a) and 4(b). It is found that of the overall reduction of J_c with α , a significant portion originates from $d_{x^2-y^2}$ symmetry and faceting. For example, of the experimentally observed reduction of J_c for a symmetric 36.8° boundary by three orders of magnitude, roughly one order of magnitude is



FIG. 6. Critical current I_c as a function of applied magnetic field H_a applied parallel to the \vec{c} -axis direction for a 16- μ m-wide and 22-nm-thick bridge across an asymmetric 45° [001] tilt YBa₂Cu₃O_{7-x} grain boundary (α_1 =45°, α_2 =0) at 4.2 K. The presented I_c is the average of the I_c values obtained with positive and negative current bias, using a 10- μ V voltage criterion.

attributed to faceting and symmetry effects. It is obvious that J_c is also strongly reduced by other mechanisms, such as disorder effects or deviations from bulk stoichiometry.^{23,24}

From the previous it is clear that straight grain boundaries of different configurations are predicted to have different J_c values for a given misalignment angle. Likewise, symmetric and asymmetric boundaries are expected to differ in behavior. Therefore, aside from the value of α and the type of misalignment, the respective α_1 and α_2 values are also relevant for the grain boundary transport properties. Although the differences between the different grain boundary configurations are expected to be quite large, they may, in a practical experiment, be masked by faceting effects.

After having discussed the influence of $d_{x^2-y^2}$ symmetry and faceting on the $J_c(\alpha)$ dependence, we now turn to their influence on the magnetic field dependence of the grain boundary critical current $I_c(H_a)$. A boundary with a highly inhomogeneous $J_c(\vec{r})$ will display strong deviations from a Fraunhofer-type $I_c(H_a)$ characteristic, the hallmark of homogeneous, small Josephson junctions. For boundaries with small misorientation angles, such as the widely used symmetric 24° [001] tilt boundary, faceting and $d_{x^2-y^2}$ symmetry may cause minor distortions of the $I_c(H_a)$ curves. For a symmetric 36.8° [001] tilt grain boundary junction the $I_c(H_a)$ dependence commonly shows nonzero lower-order minima, which we attribute to faceting and $d_{x^2-y^2}$ symmetry. The effects will become more prominent with increasing α , in particular if α_1 or α_2 is close to 45°. Indeed, for asymmetric 45° [001] tilt boundaries ($\alpha_1 = 0^\circ$, $\alpha_2 = 45^\circ$), the $I_c(H)$ dependence deviates notably from a Fraunhofer pattern due to the mechanisms described in Refs. 60-64. This was noted by Humphreys *et al.*⁶⁵ and analyzed in detail by Copetti *et al.*⁶⁶ and by us.⁶⁷ An example of such a non-Fraunhofer-like $I_c(H)$ characteristic is shown in Fig. 6 for a 16- μ m-wide asymmetric 45° [001] tilt grain boundary junction ($\alpha_1 = 0^\circ$, $\alpha_2 = 45^\circ$) patterned in a YBa₂Cu₃O_{7-x} thin film with a thickness of 22 nm. We attribute the sharp increase of I_c at $H_a \approx 13$ G to a certain degree of regularity by which YBa₂Cu₃O_{7-x} growth spirals are apparently arranged at the boundary, leading to an array of 0 and π facets. Similar peaks were observed for other samples with comparable dimensions and film thicknesses. The magnetic field dependence of the critical current for asymmetric 45° [001] tilt grain boundaries ($\alpha_1 = 45^\circ, \alpha_2 = 0^\circ$) provides a very convenient tool to distinguish between *s*-type superconductors [showing a Fraunhofer-like $I_c(H_a)$ dependence] and superconductors with other symmetry properties of the order parameter (showing a behavior as presented in Fig. 6).

Humphreys et al.⁶⁵ suggested that the distorted $I_c(H_a)$ dependence arises from magnetic flux appearing spontaneously at the intersections of the facets. Copetti et al.⁶⁶ analyzed theoretically the behavior of rings containing only one pair of 0 and π junctions. They concluded that these rings spontaneously generate magnetic flux if the absolute values of the critical currents of the two junctions are equal. If this is not the case, the flux is only generated if the inductance is sufficiently large, as is, for example, the case in the tricrystal ring experiments.¹ In the small inductance limit, the ring is expected to generate no flux. Although this limit applies to intragrain boundary effects, we anticipate spontaneous current flow and magnetic flux to appear due to the complexity of the grain boundary, particularly for high-angle grain boundaries. The magnetic flux, which may cause a weak paramagnetism of the boundary, will be unquantized. It may appear throughout the boundary, will be of alternating sign, and will depend on $J_c(\vec{r})$ and on the effective inductance of the current paths. It may be enhanced at defects such as holes or precipitates. If magnetic flux occurs spontaneously in the boundary, the current-phase relation of the grain boundary junction will deviate from the ideal sinusoidal behavior. This may lead to increased noise in the junction, particularly lowfrequency 1/f noise. The effects discussed may also affect phase-slip processes, which, e.g., determine the zero resistance critical temperature T_{c0} of the grain boundary junction.

Until now, only supercurrents across faceted grain boundaries have been discussed. In the following, we will also take the normal state transport properties into consideration. The normal state resistivity $\rho_n(\vec{r})$ is not affected by the symmetry of the superconducting order parameter. It is, however, a function of the topology of the boundary and will therefore be spatially modulated by the faceting in a way only partly correlated to the modulation of J_c . It is clear that, in addition to J_c , the $J_c \rho_n(\vec{r})$ product will also vary along the grain boundary. This variation of $J_c \rho_n$ leads to a smoothing of structures in the current-voltage characteristics that are associated with the energy gap of the superconductor. The $J_c \rho_n$ product, measured for a practical grain boundary junction, is not a direct measure of the microscopic $J_c \rho_n$ and does not necessarily characterize properties intrinsic to the junction barrier. For example, the grain boundary $J_c \rho_n$ will be reduced by (110) facets, with $J_c = 0$, and by π facets, with their negative J_c values. Evidently, the intrinsic $J_c \rho_n(J_c)$ dependence may deviate from the experimentally observed scaling law $J_c \rho_n \propto (J_c)^{0.6}$.

Owing to the mechanisms described above, the supercurrent and the normal current will be distributed differently over the grain boundary. This reduces, for example, the correlation observed between the critical current noise and the resistance noise. In accordance with experimental observations,^{68,69} this correlation is expected to decrease with increasing grain boundary angle, due to the enhanced spatial inhomogeneity of $J_c(\vec{r})$. The different, inhomogeneous, spatial distributions of the supercurrent and the normal current will affect the shape of the current-voltage characteristic of the grain boundaries and the appearance of resonances, either self-induced or induced by external rf irradiation.

In conclusion, we have analyzed the effects of $d_{x^2-y^2}$ symmetry and faceting on the transport properties of high- T_c grain boundaries. These effects account for a considerable part of the experimentally observed reduction of the critical current density J_c with increasing grain boundary angle. We pointed out that the intrinsic angle dependence of J_c is not yet known and may deviate considerably from the angle dependence observed for multifaceted grain boundary junctions. Owing to the combined effects of faceting and $d_{x^2-y^2}$ symmetry, the $J_c \rho_n$ product measured for a macroscopic grain boundary junction may differ notably from the microscopic value and is therefore not a direct measure of the intrinsic barrier properties. This also holds for the experimentally observed scaling behavior of $J_c \rho_n$ with J_c . The faceting and $d_{x^2-y^2}$ symmetry may lead to additional effects that originate from the inhomogeneous current distribution and from the difference in these distributions between the superconducting and the normal states. The experimental data presented here apply to $YBa_2Cu_3O_{7-x}$. For other high- T_c cuprates the qualitative behavior will be similar, but may be quantitatively different. The considerations presented here may help explain unexpected effects that can occur in experiments based on grain boundary junctions to probe the symmetry properties of the order parameter of high- T_c superconductors.

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